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In this report, we present our observation of optically excited nonequilibrium spin dynamics in (Ga,Mn)As at room temperature. Our observation demonstrates two different regimes: 1) an initial ultrafast oscillatory behavior during the subpicosecond and picosecond time scale after photo-excitation, which we find to be related to photo-induced carrier (electrons and holes) spins; 2) a slow demagnetization over hundreds of picoseconds, which, we believe, results from not only strong-coupled hole-Mn spins but also interactions between electron spins and Mn moments.

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Technical report

The magnetic semiconductor (Ga,Mn)As has received considerable interest in recent years, and is expected to be the material of choice for future spin-based devices or spintronics. Exploring and understanding the dynamics of spins in this material is essential for spintronics applications [1,2]. Ultrafast pump-probe magneto-optical spectroscopy opens a unique way to study time-resolved photo-induced magnetization dynamics [3,4,5]. Kojima et al. observed a magnetization relaxation on the order of 1 ns at 95K [3]. Munekata's group reported the dynamics of photoinduced magnetization rotation in ferromagnetic (Ga,Mn)As, and found that holes and Mn spins rotate and relax in tens of picoseconds after the optical excitation below transient temperature [4]. Kimel et al. have performed the time-resolved MOKE experiment at low temperature (10K and 60K) [5] and found that the photo-induced magneto-optical signal only comes from the spin-polarization decay in conduction band electrons, and no noticeable connection with Mn precession. Theoretical studies have also been done in detail recently [6,7], and an ultrafast light-induced precession and relaxation of collective magnetization is predicted. However, up to now, no time-resolved analysis of the photoinduced magnetization at room temperature was reported.

In this report, we present our observation of optically excited nonequilibrium spin dynamics in (Ga,Mn)As at room temperature. Our observation demonstrates two different regimes: 1) an initial ultrafast oscillatory behavior during the subpicosecond and picosecond time scale after photo-excitation, which we find to be related to photo-induced carrier (electrons and holes) spins; 2) a slow demagnetization over hundreds of picoseconds, which, we believe, results from not only strong-coupled hole-Mn spins but also interactions between electron spins and Mn moments.

We mainly investigated the ferromagnetic $Ga_{0.95}Mn_{0.05}As$ (300 nm) sample, which was grown by low temperature molecular beam epitaxy (LT-MBE) on GaAs/GaAs (001) substrate at substrate temperature of 275^{o}C. For comparison, a low temperature grown GaAs sample with thickness 800nm was also measured.

Time-resolved magneto-optical Kerr measurements were performed, using a Coherent MIRA 900 Ti:Sapphire laser, which produced ~150-fs-wide pulses in the 740nm (1.675eV) to 890nm (1.393eV) wavelength range with a repetition rate of 76 MHz. The pump beam was modulated between left and right-circular polarized states employing a quarter-wave plate. Probe beam is p-polarized. The rotation angle θ_F is detected using a balanced photodiode bridge, in combination with a lock-in amplifier. Both pump and probe beams are focused on the sample with a spot diameter of about 100 μm, with energy ratio of 10:1. Typical pump light has an average power of 20 mW, equivalent to a pulse energy of 0.27 nJ and a fluence of 3.4 μJ/cm^2. The pump beam is incident normal on the sample while the probe is at 45^{o}.

The magneto-optical Kerr effect (MOKE) occurs when the linear polarized light passes through a medium with birefringence, which happens when an external magnetic field or

a circularly polarized pump light induces magnetization in this medium. Therefore the change of probe polarization can be used to measure the magnetization dynamics [5,8].

In Fig.1, the measured time-resolved MOKE signals at several wavelengths in $Ga_{0.95}Mn_{0.05}As$ are shown. We noticed that polar Kerr rotation signal has a fast sub-piconsecond transient followed by a slower relaxation process on the order of 100ps. Fig.2(a) illustrates the first fast dynamics captured from Fig.1 at several wavelengths. For comparison, the Kerr rotation signals in LT-GaAs are also presented in Fig.2(b). As can be seen from Fig.2(a) and (b), for both samples the initial transients happen within 1 ps. However, in $Ga_{0.95}Mn_{0.05}As$, a picosecond relaxation is visible after the initial fast transient. We calculated the time constants following the fitting equation defined in [9]:

$$\theta_k = A\exp\left(-\frac{t^2}{4w^2}\right) + \frac{B}{2}\exp\left(\frac{w^2}{\tau_1^2} - \frac{t}{\tau_1}\right)\left[1 - erf\left(\frac{w}{\tau_1} - \frac{t}{2w}\right)\right]$$
$$\frac{C}{2}\exp\left(\frac{w^2}{\tau_2^2} - \frac{t}{\tau_2}\right)\left[1 - erf\left(\frac{w}{\tau_2} - \frac{t}{2w}\right)\right] + D \qquad (1)$$

where w is the rms width of the optical pulse, t is delay time between pump and probe pulses, τ_1 and τ_2 represent 2 relaxing time constants. Results at wavelength 840 nm for $Ga_{0.95}Mn_{0.05}As$ demonstrate that τ_1 and τ_2 have relaxation time of 50 fs and 3 ps, respectively. The initial fast transient behaviors of Kerr rotation for $Ga_{0.95}Mn_{0.05}As$ and LT-GaAs may have the same origin. As we know, a very fast carrier relaxation takes place after photo-excitation in LT-GaAs, due to the trapping process [10], which also contributes to spin relaxation of polarized electrons. Similar results have been presented in GaAs at room temperatures by Kimel et al. [9]. Because of strong spin-orbital coupling in the valence band, there exist close correlations between the momentum scattering (on the order of 10 fs [11]) and spin relaxation. And considering redistribution of carriers via hole-hole and electron-phonon scattering [2], we can assume a fast spin relaxation time due to holes. Both contributions may result in ultrafast oscillations superimposing on a steep background in $Ga_{0.95}Mn_{0.05}As$ at room temperature. The final long lasting relaxation processes are found to decay on the order of 100ps. This phenomenon cannot be associated to hole spins, which are known to relax in less than hundreds of femtoseconds[12]. The spin relaxation time of electrons in bulk undoped GaAs is also less than 10ps at room temperature [13]. Therefore, this long-lived process may be associated with spin-polarization decay of Mn ions. We noticed that the long recovery processes does not appear instantaneously when spin-polarized carriers are excited but presents itself after a gradual process within ~15ps. In view of rate equations, we cannot assume rotation of Mn spins is only caused by the photogenerated spin-polarized holes, which will only manifest an instantaneous response in Kerr rotation but not a gradual process as shown in experiment. This leads us to the hypothesis that electron spins together with hole spins trigger the rotation of Mn moments, or an interaction exists between spins of the conduction band electrons and spins of the Mn ions.

In conclusion, we carried out the first time-resolved MOKE studies in paramagnetic GaMnAs epilayers at room temperature. An ultrafast spin dynamics in subpicosecond and picosecond regime is attributed to both photogenerated holes and electrons spins. The

long-lived photoinduced magnetization rotation characterized by a relaxation time ~100 ps is found to be related to both the strong coupled hole-Mn spins and noticeable interaction between electron and Mn spins.

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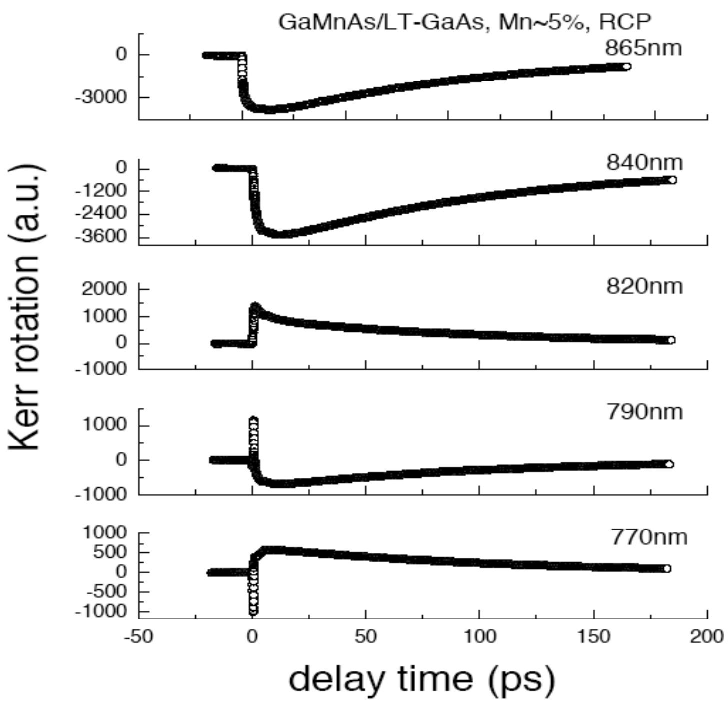
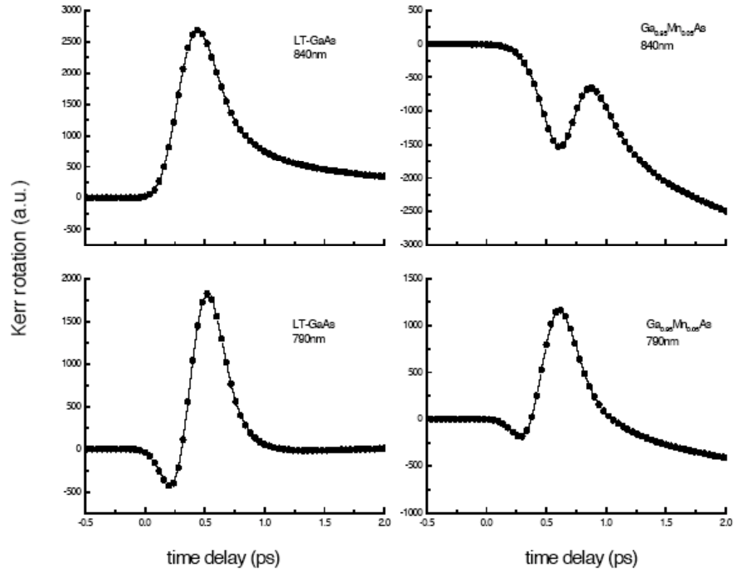


Fig.1 Dynamics of Kerr rotation in $Ga_{0.95}Mn_{0.05}As$ for different wavelengths at room temperature.



(a) (b)

Fig.2 Short-term dynamics of Kerr rotation in LT-GaAs (a) and $Ga_{0.95}Mn_{0.05}As$ (b) at different wavelengths.